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Life Cycle Assessment of Hydrogen Production Methods – A Review

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Life Cycle Assessment of Hydrogen Production Methods – A Review

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Executive Summary

The need for a sustainable energy supply is becoming more important with declining fossil energy resources, environmental pollution and climate change. Hydrogen could consequently play such a role of clean and efficient fuel, provided its production is also environment friendly. It can be produced using diverse resources including nuclear; natural gas and coal; and biomass and renewables including solar, wind, hydroelectric, or geothermal energy. As of today, the feedstock for hydrogen production is dominated by fossil fuels and the most widely used method is steam methane reforming. These methods are associated with adverse environmental impacts. Therefore, research is on-going to find the environmental friendly hydrogen production methods and feedstocks. Electrolytic hydrogen production using water as feedstock and renewable resources generated electricity as energy source could be one of the best options to meet these needs. However, this technology has yet to be cost efficient and commercial to produce hydrogen for energetic uses.

This study aims to carry out a literature review on life cycle assessment (LCA) of hydrogen production methods with focus on electrolytic hydrogen production using renewable resources generated electricity. In order to compare the results, other conventional methods of hydrogen production beyond the electrolysis have also been included in the review. This will give a comparative overview on environmental performances of all these methods. Altogether twenty-one studies (mainly journal articles and few organizational reports) that discuss on LCA of hydrogen production have been reviewed. Detail of the review has been presented in chapter IV.

Because of the differences on studies' objectives and thereby differences in system boundaries, functional units, goal and scope, etc. of individual study, it was difficult to make a direct comparison of one technology to the other. However, an attempt has been made to compare the aggregated LCA results for different non electrolytic technologies with that of electrolysis. From an environmental analysis perspective, it can be concluded that electrolytic hydrogen production using wind or hydropower generated electricity is one of the best methods for hydrogen production over that from natural gas reforming or electrolysis using the electricity from fossil fuel dominated grids.

Keywords

Alkaline electrolysis, wind electrolysis, GWP, LCA, energy carrier, hydrogen production

Contribution to ElyGrid Project

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I   Introduction

Global energy consumption is expected to increase dramatically in the next decades, driven by rising standards of living and a growing population worldwide. The increased need for more energy will require enormous growth in energy generation capacity and more secure and diversified energy sources [IEA, 2011, USDOE, 2009]. The need for a sustainable energy supply is becoming more important in light of declining fossil energy resources, environmental pollution, climate change and increasing dependency on fossil fuels exporting countries. Consequently, alternative fuels are required to fulfil criteria such as less release of carbon dioxide, sustainable resources use, suitability for the mobile (transportation) sector and being at an affordable price range. Hydrogen could play such a role of clean and efficient fuel [Romagnoli et al., 2011] when it is produced from sustainable energy resources.

Hydrogen is the simplest element on the earth. Although abundant on earth as an element, hydrogen combines readily with other elements and is almost always found as part of another substance, such as water, hydrocarbons, etc. Like electricity, hydrogen is an energy carrier and not a primary energy resource. The average energy content is about 3 kWh/Nm^3 (normal cubic meter (Nm^3) being a metric unit for quantifying hydrogen volume at 0°C and 1 atm pressure [NEEDS, 2008]). Hydrogen can be produced using fossil fuel feedstocks such as natural gas and coal as well as biomass; and also via electrolysis using the electricity from e.g. solar, wind, hydro-electric, geothermal energy or fossil fuel and nuclear dominated conventional grids. This diversity of these sources makes hydrogen a promising energy carrier in future [USDOE, 2012].

Although hydrogen is used in a number of industrial applications, with today's largest consumer being ammonia production, its energetic use is negligible. Global hydrogen production in 2001 was about 500 billion Nm^3/yr [Saur, 2008]. Ammonia production accounted for 62.4% of the world's hydrogen, and refining and methanol production consumed 24.3% and 8.7%, respectively. The rest uses include metal production, electronics manufacture, float gas production, cooling of thermal generators, etc. Because such large quantities of hydrogen are required in these instances, the hydrogen is generally produced at the consumer site, and the most common method is steam reforming of natural gas. Globally, about 96% of hydrogen production comes from fossil fuels (in the United States, about 95% of hydrogen is produced from natural gas) [Spath & Mann, 2001]. This trend has not yet altered much.

Fossil fuel combustion causes adverse environmental consequences that have motivated research on more environmentally benign alternative fuels. Hydrogen can be produced from renewable energy systems [Cetinkaya et al., 2012]. Hydrogen has been proposed as one of the potential energy carriers for future energy scenarios because several advantages are achieved when it is used: high yields in fuel cells, clean combustion without emissions of CO_2, NO_x, or SO_x, and feasible storage of the intermittent renewable energy sources [Muradov & Veziroglu, 2008]; [Balat, 2008]. Although it might be obtained from renewable energy in the long term, the fossil fuels are the most realistic alternatives for mass production in the short term [Dufour et al., 2011]. This is also clear from the figures that the global

sources of hydrogen in 2006 was about 48% from natural gas, 30% from oil, 18% from coal and only 4% by electricity via water electrolysis [PE International, 2010]. This share will not drastically change in a near future, though the most abundantly available fossil fuel coal may play a bigger role than natural gas. Electrolytic hydrogen production might lay its focus on electrolysis using the renewable resources generated electricity.

This study aims to analyse the environmental effects of different hydrogen production routes with the help of published literatures using the life cycle assessment (LCA) approach. The focus hereby is set for water electrolysis using electricity from renewable energy resources for different electrolytic hydrogen production routes (using electricity from renewable and non-renewable sources such as fossil fuel and nuclear). Their results are compared with the conventional hydrogen production routes using fossil fuels as feedstocks.

This document has been organized as follows. Section II introduces the different hydrogen production methods. As the main focus of this research, the electrolytic hydrogen production route is described in detail. Section III briefly introduces the environmental LCA. Detailed literature review on LCA of hydrogen production methods has been presented in section IV. The results are concluded in section V, which is followed by the list of references.

II   Hydrogen Production Methods

Hydrogen can be produced from a variety of feed-stocks. These include fossil resources, such as natural gas and coal, as well as renewable resources, such as biomass and water with energy input from renewable sources (e.g. solar, wind, wave or hydropower). A variety of process technologies can be used, including chemical, biological, electrolytic, photolytic and thermo-chemical. Each technology is in a different stage of development, and each offers unique opportunities, benefits and challenges. Local availability of feedstock, the maturity of the technology, market applications and demand, policy issues, and costs will all influence the choice of the various options for hydrogen production.

Several technologies are already available in the market for the industrial production of hydrogen. The first commercial technology, dating from the late 1920s, was the electrolysis of water to produce pure hydrogen. In the 1960s, the industrial production of hydrogen shifted slowly towards a fossil-based feedstock, which is the main source for hydrogen production until today [IEA, 2006]. Fig. 1 shows the main alternative methods of hydrogen production.

Fig. 1: The main alternative methods of hydrogen production
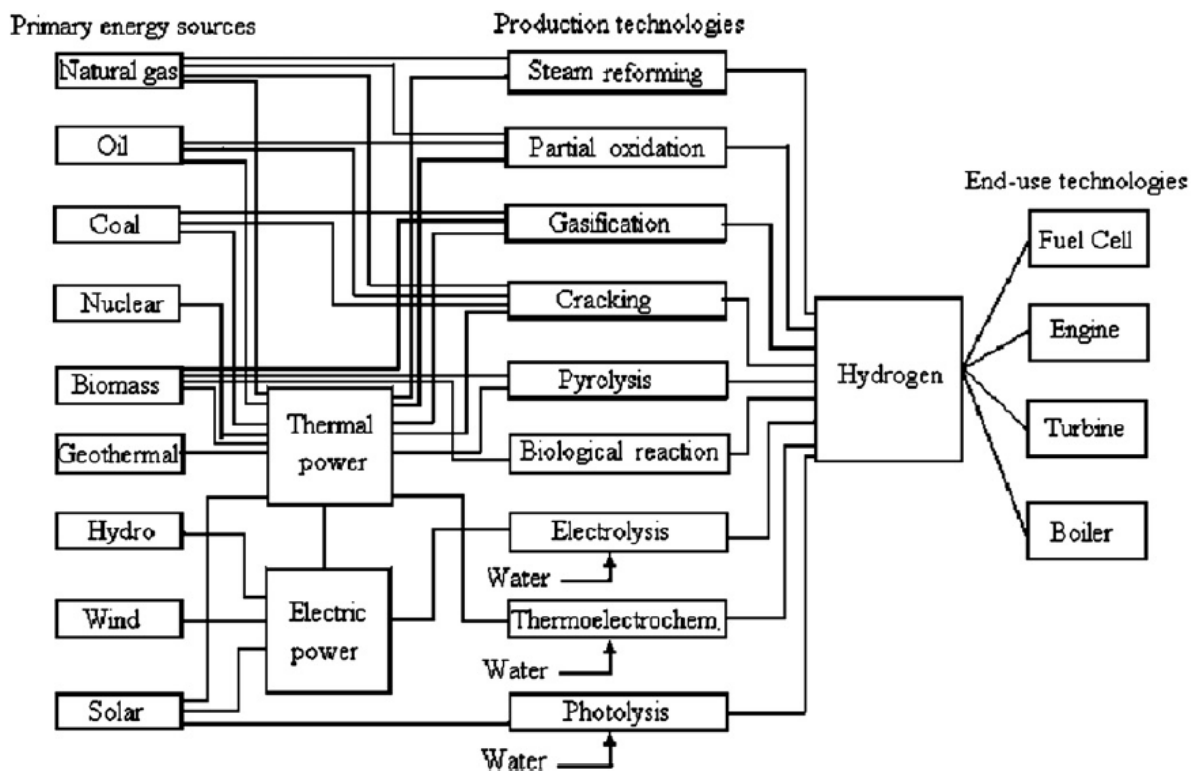
Source: [Balat, 2008]   IEK-STE 2012

Other studies already give detailed technical overview of different hydrogen production techniques, e.g. [Holladay et al., 2009] or define a "green path towards hydrogen economy", e.g. [Muradov & Veziroglu, 2008]. Also [Momirlan & Veziroglu, 2002] examined the various hydrogen production methods by outlining the economics, environmental impacts, applications, and hydrogen energy status around the world. Therefore, in the following sub-sections

this study will provide only a brief overview of these techniques for basics understanding rather than going into depth (details can be found in the studies mentioned above).

II.1 Hydrogen from fossil fuels

Hydrogen can be produced from fossil fuels. Since carbon dioxide (CO_2) is produced as a by-product, the CO_2 should be captured to ensure an environmental friendly process. The production methods from natural gas and coal are given below.

II.1.1 Production from natural gas

Hydrogen can be produced from natural gas by means of three chemical processes:

- Steam methane reforming (SMR)

- Partial oxidation (POX)

- Auto-thermal reforming (ATR)

II.1.1.1 Steam methane reforming (SMR)

Steam reforming involves the endothermic conversion of methane and water vapour into hydrogen and carbon monoxide (CO) (eq. 1). The heat is often supplied from the combustion of some of the methane feed-gas. The process typically occurs at temperatures of 700 to 850 °C and pressures of 3 to 25 bar. The product gas contains approximately 12% CO, which can be further converted to CO_2 and H_2 through the water-gas shift reaction (eq. 2). SMR constitutes roughly 50% of the global annual production of hydrogen of about 500 billion m^3 [Harrison & Ivy-Levene, 2008].

$$CH_4 + H_2O + heat \rightarrow CO + 3H_2 \qquad \text{(eq. 1)}$$

$$CO + H_2O \rightarrow CO_2 + H_2 + heat \qquad \text{(eq. 2)}$$
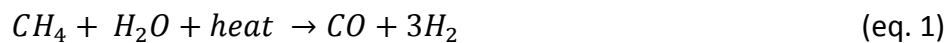
The process can be seen in a flow diagram in Fig. 2. The produced hydrogen contains impurities, e.g. CO_2 and other traces, and it should be separated. Commonly used purification process is pressure swing absorption.

Fig. 2: Hydrogen production plant
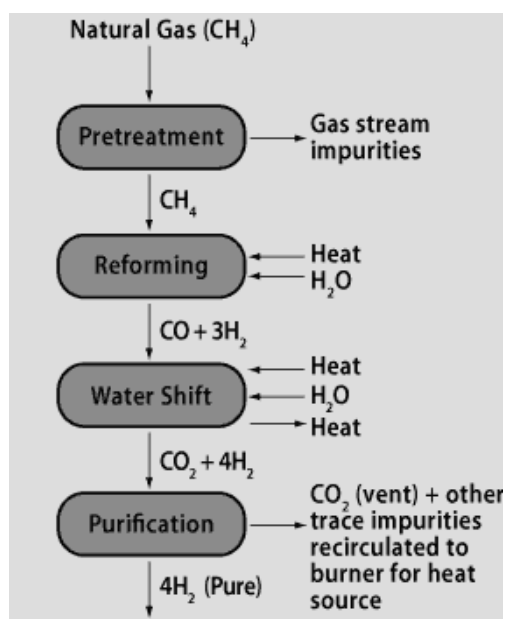
Source: Gas Technology Institute	IEK-STE 2012

II.1.1.2   Partial oxidation (POX)

Partial oxidation of natural gas is the process whereby hydrogen is produced through the partial combustion of methane (CH_4) with oxygen gas to yield CO and H_2 (eq. 3). In this process, heat is produced in an exothermic reaction, and hence a more compact design is possible as there is no need for any external heating of the reactor. Thus produced CO is further converted to H_2 as already described in (eq. 2).

$$CH_4 + \frac{1}{2}O_2 \rightarrow CO + 2H_2 + heat \qquad\qquad\text{(eq. 3)}$$

II.1.1.3   Auto-thermal reforming (ATR)

Auto-thermal reforming is a combination of both steam reforming (eq. 1) and partial oxidation (eq. 3). The total reaction is exothermic. The outlet temperature from the reactor is in the range of 950 to 1100 °C, and the gas pressure can be as high as 100 bar. Again, the CO produced is converted to H_2 through the water-gas shift reaction (eq. 2).

SMR process has several advantages such as high efficiency, less emission and less cost for large units. However, they are complex systems and are sensitive to natural gas quality. ATR or POX systems enjoy the benefits of smaller size and less costly for smaller units. However, they are less efficient, they generate more emissions and the purity of produced H_2 is less than that from SMR.

II.1.2   Production from coal

Hydrogen can be produced from coal through a variety of gasification processes (e.g. fixed bed, fluidized bed or entrained flow). Generally, high temperature entrained flow processes are favoured to maximize carbon conversion to gas, thus avoiding the formation of signifi-

cant amounts of char, tars and phenols. A typical reaction for the process is given in (eq. 4), in which carbon is converted to CO and H_2.

$$C(s) + H_2O + heat \rightarrow CO + H_2 \qquad\qquad\qquad (eq. 4)$$

Since this reaction is endothermic, additional heat is required, as with methane reforming. The CO is further converted to CO_2 and H_2 through the water-gas shift reaction as described in (eq. 2). Hydrogen production from coal is commercially mature, but it is more complex than the production of hydrogen from natural gas. The cost of the resulting hydrogen is also higher. However, as coal is plentiful in many parts of the world, it will probably be used as an energy source regardless of its other disadvantages. A process flow diagram of hydrogen production from coal gasification is given in Fig. 3.

Fig. 3: Hydrogen production via coal gasification
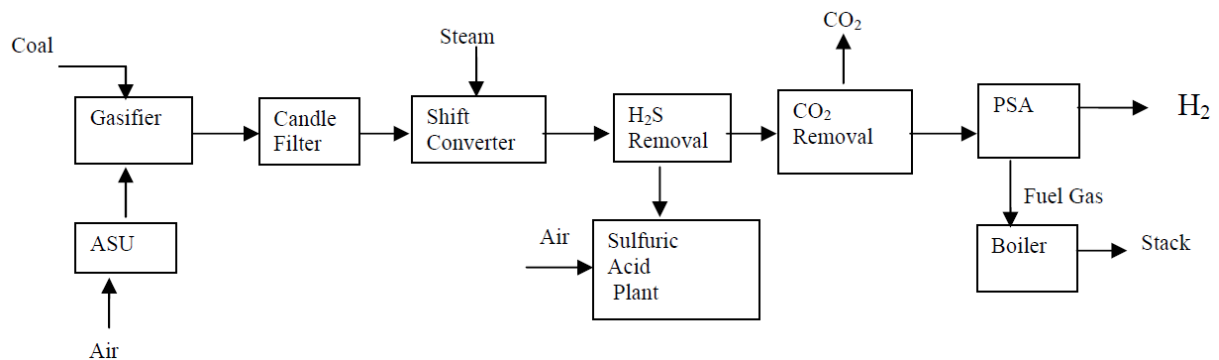
Source: [NETL, 2006]   IEK-STE 2012

II.2   Hydrogen from biomass

In biomass conversion processes, a hydrogen containing gas is normally produced in a manner similar to the gasification of coal. Currently, the pathways followed are steam gasification (direct or indirect), entrained flow gasification, and more advanced concepts such as gasification in supercritical water. Gasification and pyrolysis are considered the most promising medium-term technologies for the commercialization of H_2 production from biomass. A typical flow sheet for the production of hydrogen from biomass is presented in Fig. 4.

Fig. 4: Schematic process diagram of biomass based hydrogen production
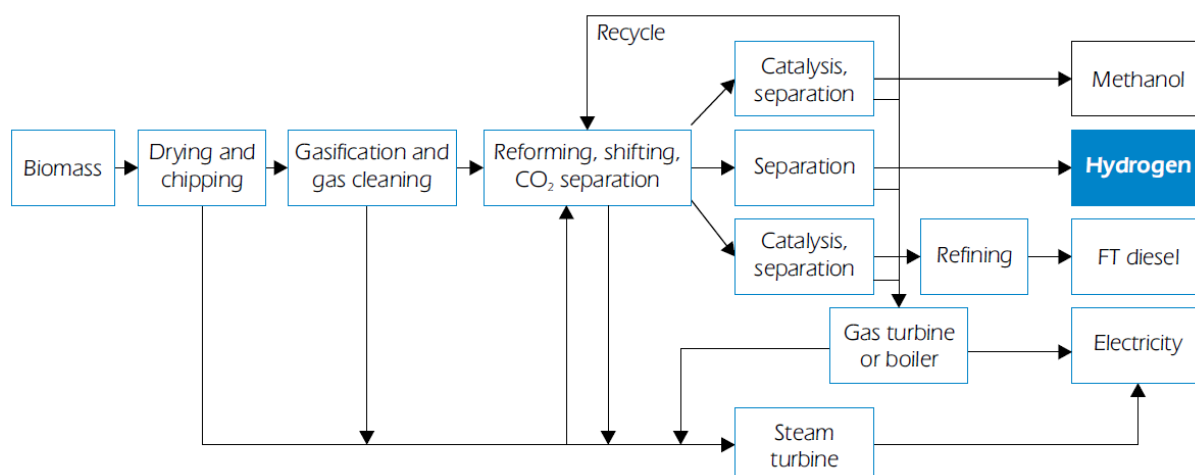
## II.3 Hydrogen from water

Hydrogen can be produced from the splitting of water through various processes such as water electrolysis, photo-electrolysis, photo-biological production and high-temperature water decomposition.

II.3.1 Photo-electrolysis (photolysis)

Photovoltaic (PV) systems coupled to electrolysers are commercially available. The systems offer some flexibility, as the output can be electricity from photovoltaic cells or hydrogen from the electrolyser. Direct photo-electrolysis represents an advanced alternative to a PV-electrolysis system by combining both processes in a single apparatus. This principle is illustrated in Fig. 5. Photo-electrolysis of water is the process whereby light is used to split water directly into hydrogen and oxygen. Such systems offer great potential for cost reduction and efficiency improvement of electrolytic hydrogen, compared with conventional two-step technologies (PV system and electrolyser). Various laboratory scale photo electrolytic cell (PEC) devices are developed, thus far demonstrating solar-to-hydrogen conversion efficiencies of up to 16% [IEA, 2006]; though even higher values up to 24% are also reported in laboratory.
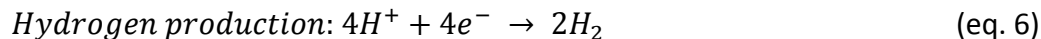
Fig. 5: Principle of photo electrolytic cell
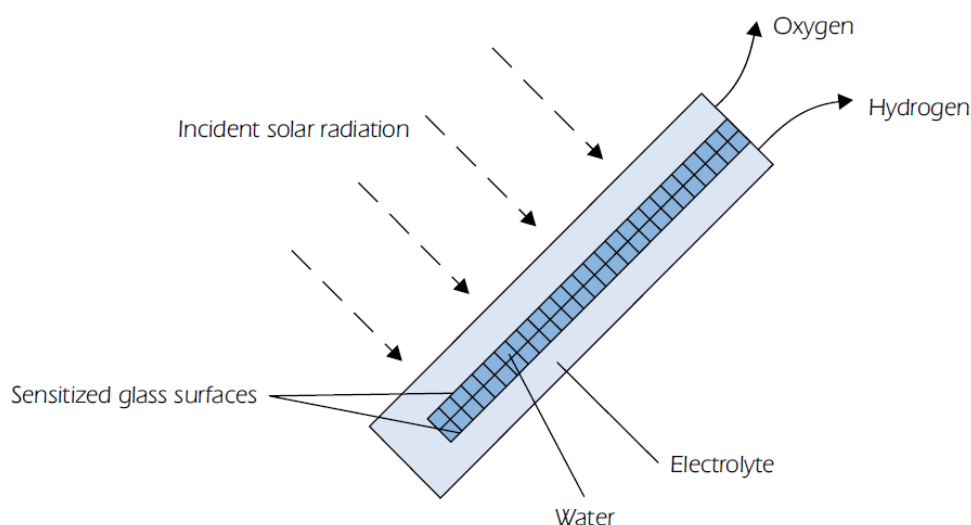
Source: Hydrogen Solar Production Company Inc.   IEK-STE 2012

II.3.2 Photo-biological production (bio-photolysis)

Photo-biological production of hydrogen is based on two steps - photosynthesis (eq. 5) and hydrogen production catalysed by algae (eq. 6). Basic principle is given in Fig. 6.

$$Photosynthesis: 2H_2O \ \rightarrow \ 4H^+ + 4e^- + \ O_2 \qquad \text{(eq. 5)}$$

$$Hydrogen \ production: 4H^+ + 4e^- \ \rightarrow \ 2H_2 \qquad \text{(eq. 6)}$$
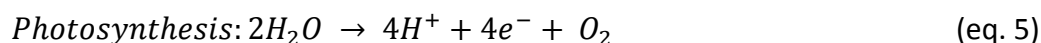
Fig. 6: Principle of photo-biological hydrogen production
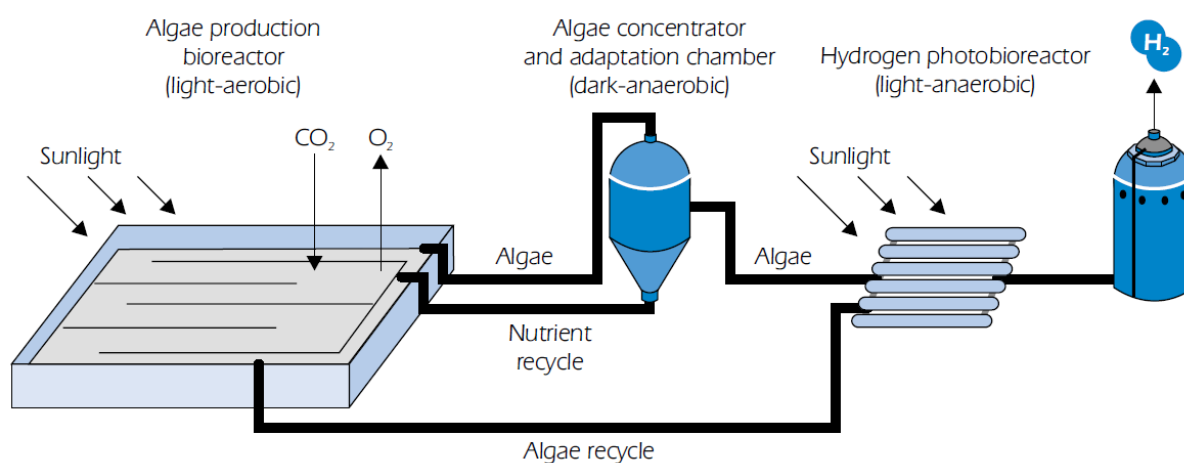
Source: [IEA, 2006]   IEK-STE 2012

II.3.3 High temperature decomposition

High temperature splitting of water occurs only at very high temperatures, e.g. about 3000 °C. At this temperature, 10% of the water is decomposed and the remaining 90% can be re-cycled. To reduce such a high operating temperature, other processes for high temperature water splitting have been developed, e.g. thermo-chemical cycles (Cu-Cl cycle, S-I cycle, etc.).

Those plants are combined with the nuclear power plants to supply this high temperature heating. Use of solar thermal or geothermal resources would also be possible.

Thermo-chemical water splitting is the conversion of water into hydrogen and oxygen by a series of thermally driven chemical reactions. While there is no question about the technical feasibility [IEA, 2006], cycles with proven low cost and high efficiency have yet to be developed commercially. An example of a thermo-chemical process is the sulphur iodine (S-I) cycle, which is shown in Fig. 7.

Fig. 7: Principle of S-I thermo-chemical process
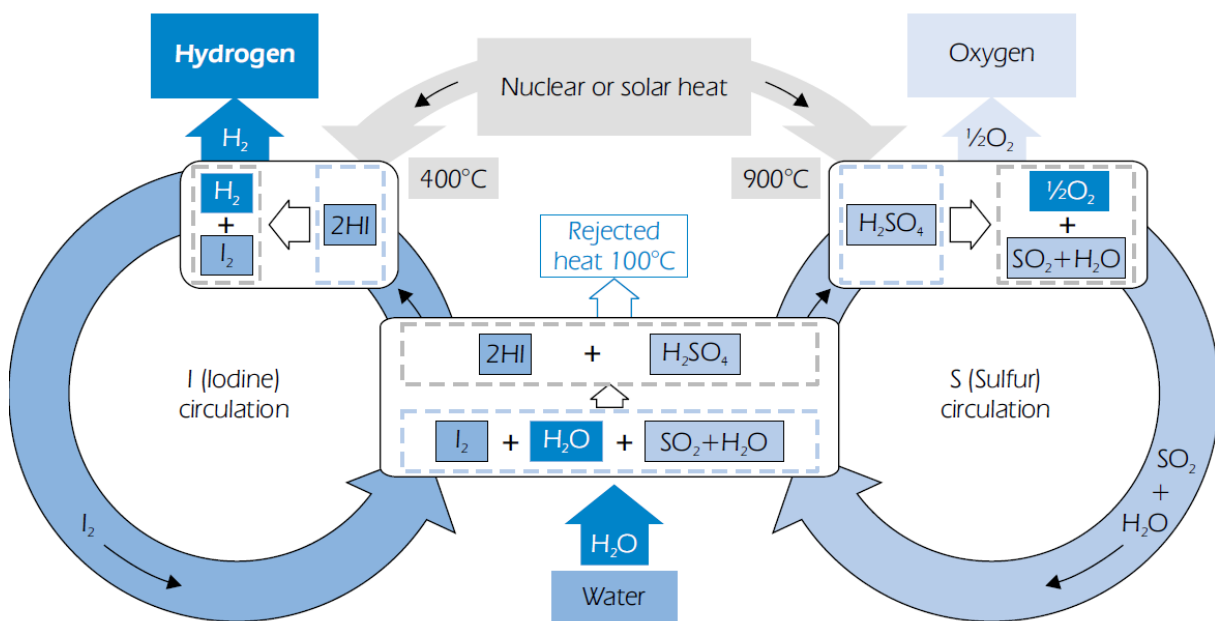
Source: [IEA, 2006]   IEK-STE 2012

For these processes, efficiencies above 50% can be expected and could possibly lead to a major decrease of hydrogen production costs. The main technical issues for these high temperature processes relate to materials development for corrosion resistance at high temperatures, high temperature membrane and separation processes, heat exchangers, and heat storage media. Design aspects for safety are also important for high temperature processes.

II.3.4 Electrolysis

Water electrolysis is a process whereby water is split into hydrogen and oxygen through the application of electrical energy (eq. 7). [Harrison & Ivy-Levene, 2008] reported that about 39 kWh of electricity and 9 litres of water are required to produce 1 kg of hydrogen at 25°C and 1 atmosphere pressure, though varying numbers can be found in the literatures.

$$H_2O + electricity \rightarrow H_2 + \frac{1}{2}O_2 \qquad\qquad (eq.\ 7)$$

The electrolysis cell is the basic element of the electrolytic hydrogen production system. The cells are connected in parallel or in series to form the electrolysis module. This is shown in Fig. 8, where V_{cell} and I_{cell} are the voltage and current of each cell, and V_M and I_M are the volt-

age and current of the module. When the electrolysis module is constituted by cells connected in parallel, the electrodes of each cell are connected to the corresponding power supply terminals. This configuration, in which each electrode has a single polarity, is named monopolar. On the other hand, when the module consists of cells connected in series, the same current flows through the cells. In this case, each electrode, except for the initial and the final ones, has two polarities, positive and negative. This configuration is named as bipolar [Ursua et al., 2012].

Fig. 8: Monopolar (a) and bipolar (b) electrolysis module
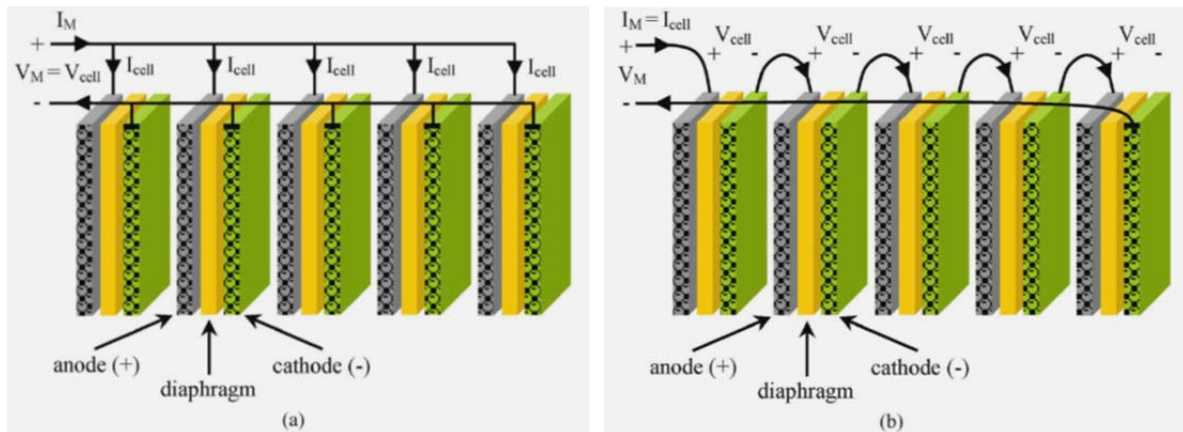
Source: [Ursua, 2010]   IEK-STE 2012

An electrolyser includes additional equipment. The hydrogen and oxygen generated are cooled, purified, compressed and stored. In many installations, the oxygen is not stored but vented to the atmosphere instead. There are also electrolysers that produce hydrogen at very high pressure, thus avoiding the compression stage and associated economic and energy cost. Water coming into the unit is previously treated so as to fulfil purity requirements to avoid mineral deposition in the cells, fast deterioration of the cell elements, and non-desired electrochemical reactions. In general, electrolysers are reliable devices that do not require continuous maintenance since they hardly include moving parts. In addition, they are silent and have a high degree of modularity, what makes them suitable for decentralized applications in residential, commercial and industrial areas.

Although electrolysers have been used for a long time, their future applications will probably often require them to be coupled to renewable energy sources in order to generate clean hydrogen and contribute to the electric grid operation. In this respect, there is still a long way to go in many aspects such as - reduction of manufacturing, distribution, and installation costs; efficiency improvement: electrolysis module, power supply, control system, etc.; operation under variable electric supply profiles; and increase of the operating temperature and pressure. There are three types of electrolysers developed so far – alkaline, polymer electrolyte and solid oxide electrolysers. They are descried below.

II.3.4.1   Alkaline water electrolysers

Alkaline water electrolysis is recognized as a mature technology; and it is more than a centu-ry old. Alkaline water electrolysers are reliable and safe, and exhibit lifetimes that can reach up to 15 years. As a result, they constitute the most extended electrolysis technology at a commercial level worldwide [Ivy, 2004]. The investment costs have been estimated to be in the range of 1000–5000 $/kW depending on the production capacity [Rajeshwar et al., 2008]. There are many manufacturers worldwide offering these systems. Most of them fab-ricate the bipolar modules. Alkaline electrolysers allow significantly efficient operation with usual values in the range of around 47 to 82% [Ursua et al., 2012].

In the last years, significant advances have been achieved regarding water alkaline electroly-sis mainly in two directions. On the one hand, the efficiency of the electrolysers has been improved with the aim of reducing operating costs associated to the consumption of elec-tricity. On the other hand, the operating current densities have been increased in order to reduce the investment costs. The investment costs are almost proportional to the electroly-sis cells surface area. Among others, these improvements include development of new ad-vanced materials to be used as diaphragms replacing the previous ones made of asbestos. In this regard, the use of ion exchange inorganic membranes has become alternative. Some examples are the membranes based on antimony polyacid impregnated with polymers, a porous composite composed of a polysulfone matrix and ZrO_2 (Zirfon), and separators based on polyphenyl sulphide (Ryton) [Vandenborre et al., 1980], [Vermeiren et al., 1998]. Also advanced alkaline water electrolysers with working temperatures up to 150 ^{o}C are devel-oped.

The operating principle of an alkaline water electrolysis cell is illustrated in Fig. 9. The cell consists of two electrodes separated by a gas-tight diaphragm. This assembly is immersed in a liquid electrolyte that is usually a highly concentrated aqueous solution of KOH of the or-der of 25–30 wt.% to maximize its ionic conductivity. Typical operating temperatures range from 65 to 100 ^{o}C. Other possible electrolytes solutions of NaOH or NaCl are less commonly used. The main and obvious drawback of the alkaline electrolyte is its corrosive character. Hydrogen gas evolves from cathode, where water is reduced yielding hydroxide anions that circulate across the diaphragm to anode within the electric field established by external power source. The hydroxide anions recombine on anode surface to produce oxygen. The following reactions take place inside an alkaline electrolysis cell:

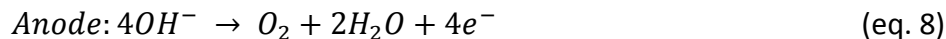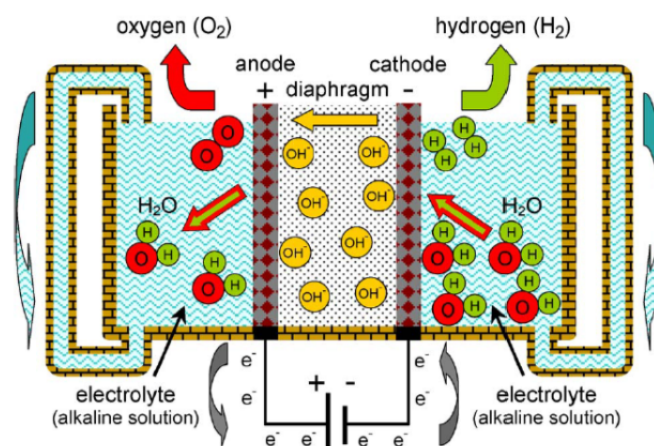$$Anode: 4OH^- \ \rightarrow \ O_2 + 2H_2O + 4e^- \qquad\qquad\qquad \text{(eq. 8)}$$

$$Cathode: 4H^+ + 4e^- \ \rightarrow \ 2H_2 \qquad\qquad\qquad\qquad \text{(eq. 9)}$$

$$Sum: 2H_2O \ \rightarrow \ O_2 + 2H_2 \qquad\qquad\qquad\qquad\quad \text{(eq. 10)}$$

Fig. 9: Operating principle of an alkaline electrolysis cell

Advanced alkaline electrolysers are the most suitable for large-scale hydrogen production. Some manufacturers (e.g. Statoil, H2 Logic, IHT etc.) fabricate units with very high production capacities, e.g. in the range of 500–760 Nm^3/h, corresponding to electric power consumptions of about 2150–3534 kW. As concerns the temperature, the operating range is typically between 5 °C and 100 °C depending on the model, although some prototypes can reach up to 400 °C [Ganley, 2009]. Regarding the electrolysis pressure, there are models operating at atmospheric pressure, whereas others can reach up to 448 bar. Nevertheless, the maximum electrolysis pressure is typically close to 25–30 bar [Ursua et al., 2012]. The purity levels of hydrogen and oxygen can reach 99.9 and 99.7 vol.%, respectively, without auxiliary purification equipment [Ivy, 2004]. On the other hand, the water fed to the electrolyser has to be significantly pure, with conductivity below 5 µS/cm, in order to protect the electrodes.

Alkaline electrolyser systems typically contain the main components as shown in Fig. 10. The major challenge is to design an electrolyser at lower cost with higher energy efficiency.

Fig. 10: Process diagram of alkaline electrolysis
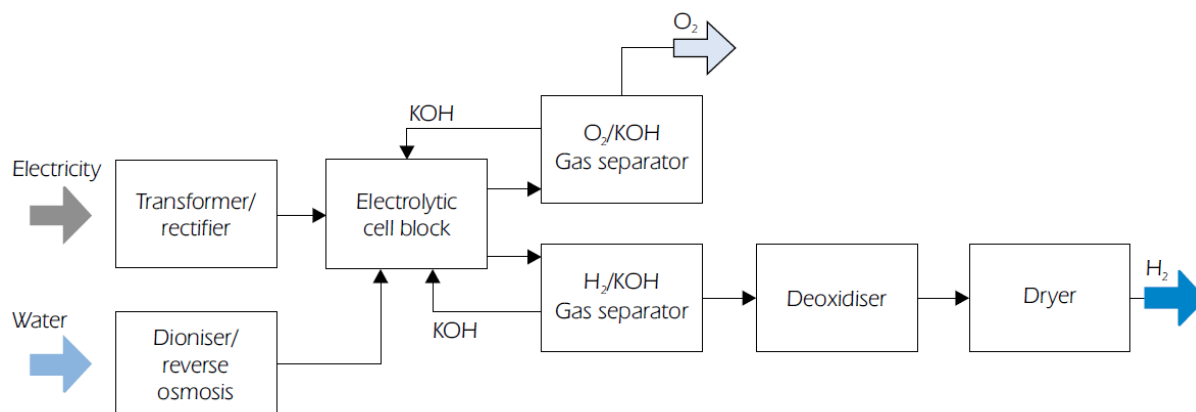
II.3.4.2   Proton exchange membrane electrolysers

This technology is also referred to as polymer electrolyte membrane (PEM) or solid polymer electrolyte (SPE). Currently, there are very few companies manufacturing PEM electrolysers. This is mainly due to their limited production capacity, short lifetime, and comparatively high investment cost. In PEM electrolysers, the electrolyte is a gas-tight thin (below 0.2 mm in thickness) polymeric membrane. As for the PEM fuel cells, the most commonly used membrane for water electrolysis is Nafion. The modules of the PEM electrolysers almost invariably adopt a bipolar configuration. Anode, cathode, and membrane set a so-called membrane electrode assembly (MEA) constitute. The electrodes typically consist of noble metals such as platinum or iridium. The operating principle of a PEM electrolysis cell is shown in Fig. 11.

$$Anode: H_2O \rightarrow \frac{1}{2}O_2 + 2H^+ + 2e^- \qquad \text{(eq. 11)}$$

$$Cathode: 2H^+ + 2e^- \rightarrow H_2 \qquad \text{(eq. 12)}$$
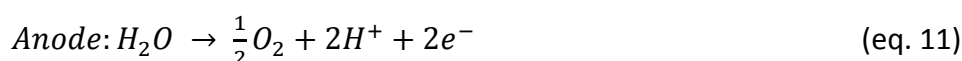
Fig. 11: Scheme of the operating principle of a PEM cell
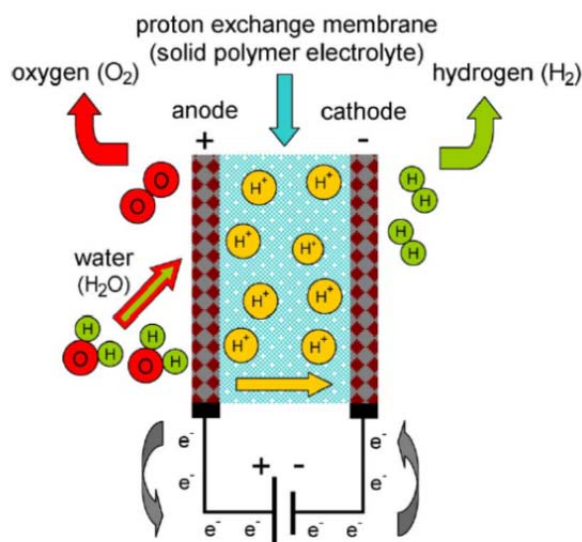
Source: [Ursua, 2010]   IEK-STE 2012

At the anode, water is oxidized to produce oxygen, electrons, and protons that circulate across the membrane to the cathode where they are reduced closing the circuit and producing hydrogen that bubbles towards the cathode gas manifold (eq. 11-12). PEM electrolysers are commercially available for low scale production applications, e.g. a model from [PROTON, 2012] has hydrogen output of 30 Nm^3/h with a power consumption of 174 kW. Their approximate efficiencies range from 48 to 65%. The presence of the polymeric membrane limits the electrolysis temperatures to values usually below 80 °C. Some models reach pressures up to 85 bar. This is due to the compact character of the electrolysis modules as well as the structural properties of the MEAs, which can endure big pressure differences between the electrode compartments. This enables, e.g., production of hydrogen at 35 bar and oxygen at atmospheric pressure. The hydrogen purity, higher than in alkaline electrolysers, is typically above 99.99 vol.% (in some cases up to 99.999 vol.%) without the need of

auxiliary purification equipment [Ursua et al., 2012]. Moreover, the very low gaseous permeability of the polymeric membranes lowers the risk of formation of flammable mixtures; hence to operate at very low current densities is permissible. The electric conductivity of the water fed to the electrolyser has to be below 1 µS/cm. PEM electrolysers have ability to work under variable power feeding regimes. This is due to the fact that the proton transport across the polymeric membrane responds quickly to power fluctuations. This is in contrast with alkaline electrolysers, where the ionic transport in liquid electrolytes shows a greater inertia [Rajeshwar et al., 2008]. Although commercially available, PEM electrolysers have some drawbacks. The main problem is perhaps their high investment costs, mainly associated to the membranes and the noble metal based electrodes. PEM electrolysers exhibit shorter lifetimes than the alkaline technology. The production capacity needs to be increased for their commercialization [Grigoriev et al., 2006].

II.3.4.3   Solid oxide electrolysers

SOEs constitute an advanced concept enabling water, or rather, steam electrolysis at high temperatures (600 - 900 ^{o}C), which results in higher efficiencies compared to alkaline or PEM electrolysers [Brisse et al., 2008]. The operating principle of a SOE cell is shown in Fig. 12. Both steam and recycled hydrogen are fed to the cathode, where water is reduced to produce hydrogen. The oxide anions generated in the cathode pass through the solid electrolyte to the anode, where they recombine forming oxygen and closing the circuit with the released electrons.

Fig. 12: Scheme of the operating principle of a SOE cell
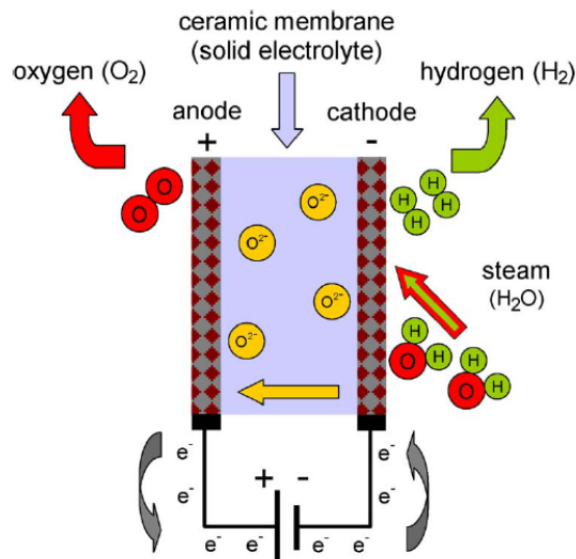
As for solid oxide fuel cells, a gas-tight thin film of yttria (Y_2O_3) stabilized zirconia (ZrO_2) (YSZ) is typically used as the solid oxide electrolyte. The cathode is a cermet usually consisting of nickel and YSZ. The anode is commonly a composite of YSZ and perovskites such as lanthanum manganites ($LaMnO_3$), ferrites ($LaFeO_3$) or cobaltites ($LaCoO_3$) partially substituted with

strontium in order to promote structural and electronic defects that increase the electro catalytic activity.

Steam electrolysis emerged with the aim of reducing the energy intensiveness and thus the operating costs of conventional water electrolysis [Salzano et al., 1985]. Theoretically up to 40% of the energy required to produce hydrogen from steam electrolysis at 1000 ^{o}C can be supplied as heat at that temperature [Brisse et al., 2008]. The features of the SOEs operation make this technology very attractive for hydrogen production when a high temperature heat source is available. This is the reason of the interest of the nuclear energy sector on steam electrolysis in view of nuclear reactors to act as a heat source at temperatures up to 950 ^{o}C. Also, the geothermal energy has potential as a heat source for steam electrolysis. Currently, SOEs are at the R & D stage. The main current obstacle for the industrial application of SOEs is the limited long term stability of the electrolytic cells. The typical specifications different electrolyzers have been summarized below.

Tab. 1: Typical specification of alkaline, PEM and solid-oxide electrolyzers

Specification	Unit	Alkaline [a]	PEM [b]	SOEs [c]
Technology maturity		State of the art	Demonstration	R & D
Cell temperature	^{o}C	60-80	50-80	900-1000
Cell pressure	bar	< 30	< 30	< 30
Current density	A/cm^2	0.2-0.4	0.6-2.0	0.3-1.0
Cell voltage	V	1.8-2.4	1.8-2.2	0.95-1.3
Power density	W/cm^2	Up to 1.0	Up to 4.4	-
Voltage efficiency	%	62-82	67-82	81-86
Specific energy consumption, system	kWh/Nm^3	4.5-7.0	4.5-7.5	2.5-3.5
Partial load range	%	20-40	0-10	-
Cell area	m^2	< 4	< 300	-
Hydrogen production, system	Nm^3/hr	< 760	< 30	-
Life time, stack	hr	< 90000	< 20000	< 40000
System life time	yr	20-30	10-20	-
Purity of hydrogen produced	%	> 99.8	99.999	-
Cold start up time	min.	15	< 15	> 60

Source: most of the data compiled and modified from [a],[b] [Smolinka et al., 2011] and [c] [NEEDS, 2008]

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II.4   Summary

Tab. 2 provides an overview on challenges and benefits of the methods described above.

Tab. 2: Hydrogen production technology overview

Technology	Challenges	Benefits
Natural gas reforming	• High capital costs • High operation and maintenance costs	• Most viable approach to begin hydrogen market in near term • Lowest current cost • Existing feedstock infrastructure
Coal and biomass gasification	• High reactor costs • System efficiency • Feedstock impurities	• Provides low-cost synthetic fuel in addition to hydrogen • Uses abundant and affordable coal feedstock
Water electrolysis	• Low system efficiency and high capital costs • Integration with renewable energy sources	• Produces virtually no pollution with renewable energy sources • Uses existing infrastructure • Uses fuel cell knowhow
Photo-electrolysis	• Effective photo catalyst material • Low system efficiency • Cost-effective reactor • Longer-term technology	• Operates at low temperatures • Clean and sustainable - using only water and solar energy
Bio-photolysis	• Efficient microorganisms for sustainable production • Optimal microorganism functionality in a single organism • Longer-term technology	• Clean and sustainable • Tolerant of diverse water conditions • Self-sustaining
Thermochemical	• Cost-effective reactor • Effective and durable materials • Longer-term technology • Nuclear issues, when nuclear fuel is a heating source	• Produces hydrogen using only water, energy from the sun or nuclear reactors, and chemicals that are recycled. • Clean and sustainable

Source: compiled from [USDOE, 2009]   IEK-STE 2012

III   Life Cycle Assessment

As environmental awareness increases, industries and businesses are assessing how their activities affect the environment. Society has become concerned about the issues of natural resource depletion and environmental degradation. The environmental performance of products and processes has become a key issue and different tools are developed to assess such performance. One such tool is life cycle assessment (LCA). The term "life cycle" refers to the major activities in the course of the product's life-span from mining, its manufacture, use to its final disposal [Curran, 2006, ISO, 2006a, ISO, 2006b].

Life cycle assessment is a "cradle-to-grave" approach for assessing industrial systems. "Cradle-to-grave" begins with the gathering of raw materials from the earth to create the product and ends at the point when all materials are returned to the environment. LCA evaluates all stages of a product's life from the perspective that they are interdependent, meaning that one operation leads to the next. By including the impacts throughout the product life cycle, LCA provides a comprehensive view of the environmental aspects of the product or process and a more accurate picture of the true environmental trade-offs in product and process selection.

Specifically, LCA is a technique to assess the environmental aspects and potential impacts associated with a product, process, or service, by:

- Compiling an inventory of relevant energy and material inputs and environmental releases

- Evaluating the potential environmental impacts associated with identified inputs and releases

- Interpreting the results to help decision-makers make a more informed decision.

LCA is an established and internationally accepted method that is defined in ISO standards: ISO 14040 [ISO, 2006a] and ISO 14044 [ISO, 2006b]. The LCA process is a systematic, phased approach and consists of four components: goal and scope definition, inventory analysis, impact assessment, and interpretation as illustrated in Fig. 13.

Fig. 13: Methodology of LCA
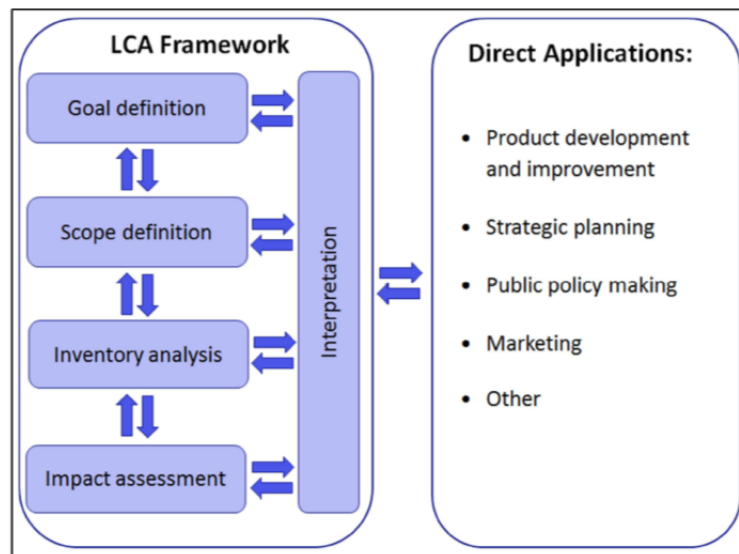
Source: [ISO, 2006a], [FC HyGuide, 2011]   IEK-STE 2012

III.1   Goal and scope definition

During goal definition the purpose of the analysis and methods for life cycle environmental impacts into the decision-making process are defined. In scope definition the investigated system is described and the bases for comparison (functional unit) as well as system boundaries concerning time frame and region are defined.

III.2   Inventory analysis

A life cycle inventory (LCI) is a process of quantifying energy and raw material requirements, atmospheric emissions, waterborne emissions, solid wastes, and other releases for the entire life cycle of a product. In LCI, all relevant data is collected on a single process scale and organized.

III.3   Life cycle impact assessment (LCIA)

In the life cycle impact assessment (LCIA) the gathered and aggregated inputs and outputs of the system are categorized and allocated to impact categories such as global warming potential, acidification potential, photochemical ozone creation potential, eutrophication potential, human toxicity potential, etc.

The Center of Environmental Science of Leiden University (CML) published an "operational guide to the ISO standards" in 2001 [Guinee, 2001], which describes the LCA procedures according to ISO standards. A set of impact categories and the characterization methods and factors for an extensive list of substances (resources from/emissions to nature) are recommended for the impact assessment phase of LCA. The environmental impact categories used in this study and their definitions are explained as follows:

III.3.1.1 Acidification potential

Acidification potential (AP) is related to atmospheric deposition by hydrogen ion concentration of acidifying pollutants on soil, groundwater, surface waters, biological organisms, ecosystems and materials. SO_2, NO_x and NH_x are the major acidifying pollutants. Unit of indicator for this impact category is kg SO_2-eq.

III.3.1.2 Eutrophication potential

Nutrient enrichment may cause an undesirable shift in species composition and elevated biomass production in both aquatic and terrestrial ecosystems. Eutrophication covers all potential impacts of excessively high environmental levels of macronutrients, mainly nitrogen and phosphorus. All emissions, which have similar effects, are also treated under the impact category eutrophication potential (EP). Unit of indicator for EP is kg PO_4^{3}-eq.

III.3.1.3 Global warming potential

Global warming potential (GWP) is defined as the impact of human emissions on the radiative forcing (i.e. heat radiation absorption) of the atmosphere, which causes temperature rise at the earth's surface. This is popularly referred as greenhouse effect. Unit of indicator for GWP is kg CO_2-eq.

III.3.1.4 Ozone depletion potential

The thinning of the stratospheric ozone layer as a result of emissions is defined as stratospheric ozone depletion. The thinning causes a greater fraction of solar ultraviolet radiation to reach the earth's surface, which has potentially harmful impacts on human health, animal health, terrestrial and aquatic ecosystems, biochemical cycles and materials. Unit of indicator for ODP is kg CFC-11 (R_{11})-eq.

III.3.1.5 Photochemical ozone creation potential

Photo-oxidant formation is the formation of reactive chemical compounds such as ozone by the action of sunlight on certain primary air pollutants. These reactive compounds may cause serious human health problems. Photo-oxidants may be formed in the troposphere under the influence of ultraviolet light, through photochemical oxidation of Volatile Organic Compounds (VOCs) and carbon monoxide in the presence of nitrogen oxides. The most important of these oxidizing compounds is ozone. Unit indicator for Photochemical Ozone Creation Potential (POCP) is kg Ethylene-eq.

III.3.1.6 Radiation

The impact category "radiation (RAD)" comprises the impacts arising from releases of radioactive substances as well as direct exposure to radiation, such as building materials, and this is harmful to both human beings and animals. The radioactivity of substance is expressed in Becquerel (Bq) per kg and category indicator for this impact category is disability adjusted life years (DALY).

Some approaches combine the different impact categories into one single score value. Because of its dependence on value judgments, weighting in LCA is a controversial issue. It is often difficult to estimate the overall environmental impact of a product. Several weighting methods have been presented to assist life cycle impact assessment, e.g. eco-indicator 95/99, EPS 2000, IMPACT 2002$^+$, or TRACI. The results are often questioned because these methods are also based on principles selected by inventors of this method. Nevertheless, these methods provide an additional set of insights to better understand environmental impact.

III.4 Life cycle interpretation

This step summarizes the results from inventory analysis and impact assessment. The outcome is a set of conclusions, recommendations and limitations.

An LCA can help decision makers select the product or process that result in the least impact to the environment. This information can be used with other factors, such as cost and performance data to select a product or process.

IV   Literature Review on the LCA of Hydrogen Production

Hydrogen can be produced from a range of technologies, the most mature of which are steam reformation of methane gas, gasification and electrolysis. As mentioned in section I, global hydrogen production in 2006 was about 48% from natural gas, 30% from oil, 18% from coal and only 4% by electricity via water electrolysis [PE International, 2010]. Electrolysis is the most important method to obtain hydrogen from water. The hydrogen obtained with this technology has a high purity that can reach to 99.999% by volume. In natural gas steam methane reforming process, the output gas from a pressure swing absorber (PSA) treatment unit is purified to 99.6% [NETL, 2006]. Further purification steps are needed to obtain higher purity level gas. Since electrolytic hydrogen is suitable for being directly used in low temperature fuel cells, high purity levels are of great advantages against both fossil fuels and biomass based reforming and gasification processes [Ursua et al., 2012]. Although this is the least efficient form of electrolyser, alkaline electrolysis is the most widely used [Simons & Bauer, 2011].

Although hydrogen is generally considered to be a clean fuel in its use phase, it is important to recognize that its production still has impacts on the environment. Examining the resource consumption, energy requirements, and emissions from a life cycle point of view gives a complete picture of the environmental burdens associated with hydrogen production [Spath & Mann, 2004]. The production of hydrogen is normally categorized into three phases: plant (hardware) manufacturing and its installation, plant operation to generate hydrogen (energy used to operate as well as feedstock for hydrogen), and the storage and/or delivery of the produced hydrogen. The environmental impacts associated with hydrogen production in almost all methods, i.e. from steam methane reforming to electrolysis, are mainly in the plant operation phase. In steam methane reforming, it is due to the consumption of natural gas as feedstock and in electrolysis it is due to the use of electricity to operate the electrolyser. These impacts can be minimized in electrolysis process if the hydrogen is produced using electricity from renewable energy sources such as wind energy. Wind based electrolysis process chain includes manufacturing and operation of wind turbines and electrolysers; and hydrogen compression/storage phases. During operation of such system, there are almost no emissions. However, production of wind turbines and electrolysers are mainly responsible for emissions.

A typical system boundary for the electrolytic hydrogen production system using power supply from renewable energy has been shown in Fig. 14. Outer dashed line represents the system boundary for such plant, a typical boundary used by most of the reviewed papers. Use phase of hydrogen, outside of the boundary in Fig. 14, has been included within the boundary only in few cases.

Fig. 14: LCA relevant stages for renewable energy based alkaline electrolyzer
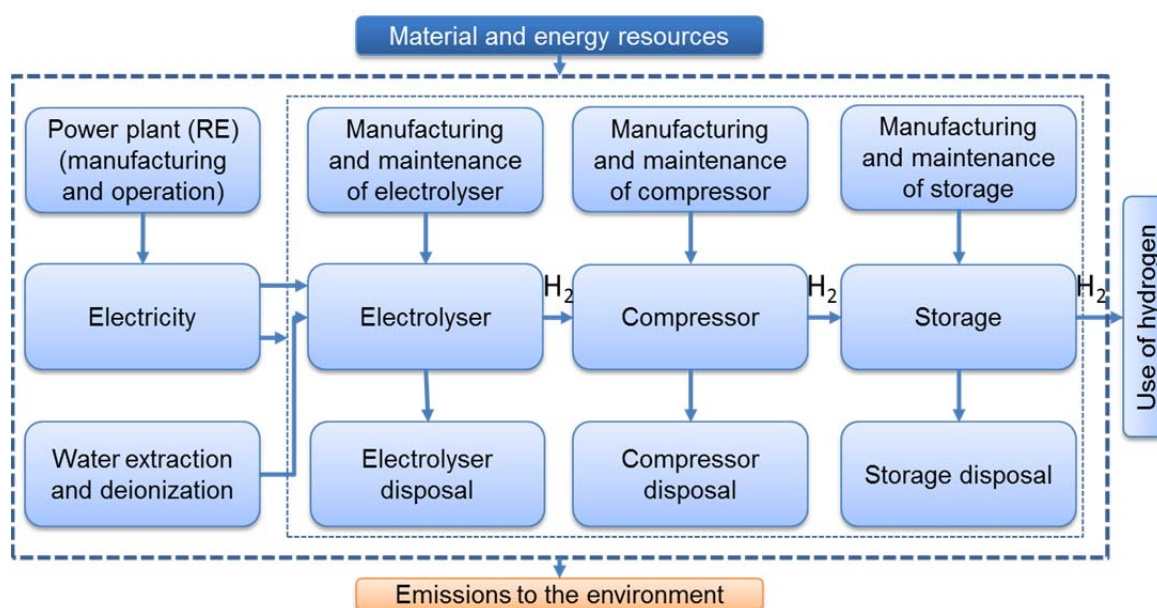
This section aims to carry out a literature review on the environmental life cycle assessment (LCA) of water electrolysis for hydrogen production. Although the study's focus is alkaline electrolysis, other types of hydrogen production processes such as thermal and thermo-chemical are also analyzed for comparison purpose. It was not possible to locate the literatures that explicitly detail the membrane materials used in electrolysers (e.g. asbestos or others) and the membrane's contribution to the environmental impacts from electrolysers. The following section firstly describes the alkaline electrolysis process chain and it is then followed by LCA review discussion.

There are several literatures that discuss the conventional hydrogen production methods using fossil fuels. However, only few discuss the environmental impacts of hydrogen production. Although in fewer numbers, literatures are also available regarding the electrolytic hydrogen production methods; however, those extending to environmental impacts of electrolytic hydrogen production are limited. The studies that were assessed as relevant to fulfill this study's aim on LCA review of hydrogen production methods are listed in Tab. 3.

Tab. 3: Reviewed literatures on LCA of hydrogen production

Authors	Title
[Boyano et al., 2011]	Exergo-environmental analysis of a steam methane reforming process for hydrogen production
[Cetinkaya et al., 2012]	Life cycle assessment of various hydrogen production methods

[Dufour et al., 2009]	Life cycle assessment of processes for hydrogen production. Environmental feasibility and reduction of greenhouse gases emissions
[Dufour et al., 2012]	Life cycle assessment of alternatives for hydrogen production from renewable and fossil sources
[Geerken et al., 2004]	Review of hydrogen LCA's for the Hysociety project
[Granovskii et al., 2006]	Life cycle assessment of hydrogen fuel cell and gasoline vehicles
[Hacatoglu et al., 2012]	Comparative life cycle assessment of hydrogen and other selected fuels
[Ivy, 2004]	Summary of electrolytic hydrogen production
[Koroneos et al., 2004]	Life cycle assessment of hydrogen fuel production processes
[Koroneos et al., 2008]	Hydrogen production via biomass gasification - A life cycle assessment approach
[Lee et al., 2010]	Life cycle environmental and economic analyses of a hydrogen station with wind energy
[Marquevich et al., 2002]	Life cycle inventory analysis of hydrogen production by the steam-reforming process: comparison between vegetable oils and fossil fuels as feedstock
[NEEDS, 2008]	Generation of the energy carrier hydrogen - In context with electricity buffering generation through fuel cells
[NETL, 2006]	Life-cycle analysis of greenhouse gas emissions for hydrogen fuel production in the United States from LNG and coal
[Ozbilen et al., 2011a]	A comparative life cycle analysis of hydrogen production via thermochemical water splitting using a Cu-Cl cycle
[Ozbilen et al., 2012b]	Life cycle assessment of hydrogen production via thermochemical water splitting using multi-step Cu–Cl cycles
[Solli et al., 2006]	Fission or fossil: Life cycle assessment of hydrogen production
[Spath & Mann, 2001]	Life cycle assessment of hydrogen production via natural gas steam reforming

[Spath & Mann, 2004]	Life cycle assessment of renewable hydrogen production via wind/electrolysis
[Utgikar & Thiesen, 2006]	Life cycle assessment of high temperature electrolysis for hydrogen production via nuclear energy
[Wulf & Kaltschmitt, 2012]	Life cycle assessment of hydrogen supply chain with special attention on hydrogen refueling stations

Source: authors' compilation   IEK-STE 2012

The following section discusses the reviewed studies individually. Because of differences in system boundary assumptions, different system sizes, different methods used for environmental impact assessment, different functional units and other several such differences, it is difficult to make an accurate and direct comparison of results from one LCA study to the other one. However, an attempt has been made to analyze the individual study at first and compare the results with each other to get a broad picture on environmental impacts ranges of different technologies at the end of this section.

[Boyano et al., 2011] have analyzed the exergo-environmental analysis of SMR process for hydrogen production. Even though their focus is on exergy analysis, they have compiled the results from eight LCA studies and presented in their paper. Seven of these studies use Eco-indicator method for impact assessment and one uses CML method. Their Eco-indicator points for electrolytic hydrogen production vary from 0.05 mPt/Nm^{3}H$_2$ for wind electrolysis to 10.30 mPt/Nm^{3}H$_2$ for biomass-gasification-electricity-electrolysis route. These values for hydropower and solar PV based electrolysis are 0.08 and 0.52 mPt/Nm^{3}H$_2$, respectively. To make a comparison, their compiled value for natural gas SMR is 0.4 mPt/Nm^{3}H$_2$, making this process better than solar PV based electrolysis system.

[Cetinkaya et al., 2012] have reported the LCA for five methods of hydrogen production: steam reforming of natural gas, coal gasification, water electrolysis via wind and solar electrolysis, and thermochemical water splitting with a Cu-Cl cycle. This paper uses secondary data for most of these discussed production methods and LCA. The system they have considered for the wind electrolysis consists of data from [Spath & Mann, 2004]. The production capacities considered for wind turbine and PV are relatively lower than that for others (Tab. 4). Between natural gas steam reforming and coal gasification based methods, the latter is more advantageous from GWP perspective. One of their figures (figure 9) indicates that the thermochemical water splitting using nuclear Cu-Cl cycle is the most environmentally benign method of hydrogen production with respect to GWP. It is followed by wind and solar PV. Indeed their table 11 shows that this Cu-Cl cycle is the worst (Tab. 4 in this study), but it is assumed to be a typing or calculation mistake because their conclusion and abstract follow the figure 9's results and not from the table. Like in the studies e.g. from [Ozbilen et al., 2011a], it is difficult to discuss the impacts of nuclear power use without commenting on the

politically sensitive issues of accidents, proliferation and spent fuel management. Tab. 4 shows a summary of hydrogen production capacity and GWP.

Tab. 4: Hydrogen production capacity and GWP values

H_2 production methods	H_2 production capacity (kgH$_2$/day)	GWP (kgCO$_2$ eq./kgH$_2$)
Steam reforming of natural gas	111,200	11.9
Coal gasification	284,000	11.3
Water electrolysis via wind energy	14	0.97
Water electrolysis via PV energy	160	2.4
Thermochemical water splitting via Cu-Cl cycle	124,800	12.3^1 $(0.7)^2$

Source: [Cetinkaya et al., 2012]   IEK-STE 2012

[Dufour et al., 2009] studied the LCA of hydrogen production via thermal and autocatalytic decomposition of methane and compared the results with steam reforming with and without carbon capture and storage (CCS). They have chosen functional unit as 1 Nm^3 of hydrogen; LCA tool as SimaPro 7.1; and impact assessment method as Eco-indicator 95. Their results show that full autocatalytic decomposition is the most environmental friendly process for hydrogen production in terms of Eco-indicator single score (about 0.24 mPt/Nm^{3}H$_2$) and also of CO$_2$ emissions. Steam reforming with CCS is far better than without CCS in terms of CO$_2$ emissions (0.31 vs 0.95 kgCO$_2$ eq./Nm^{3}H$_2$); however, surprisingly, the latter is slightly better for single score point (0.45 vs 0.47 mPt/Nm^{3}H$_2$). This is due to the fact that the CCS process requires electricity leading to more NO$_x$ emissions than conventional SMR and thus leading to higher acidification and winter smog impacts (reference has been made for World Energy Outlook electricity generation mix for year 2004). Only the use renewable electricity would make SMR with CCS attractive in this case.

In their recent study [Dufour et al., 2012] have compared GWP, CED and cumulative exergy demand (CExD) of hydrogen production from water photo-splitting, solar two-step thermochemical cycles and auto maintained methane decomposition to steam methane reforming with CCS and electrolysis via different electricity sources. The functional unit is 1 Nm^3 of hydrogen with 99.99% of purity, the inventory analysis is based on Ecoinvent database, and the

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[1] This is considered as a typo in their study, because it does not support abstract and conclusion.

[2] This value is derived from figure 9 in their paper, exact value may slightly differ.

impact assessment is based on SimaPro 7.1 software. Alkaline electrolysis process has been assessed for three electricity origin: average European electricity generation mix (33% coal, 9% hydro, 19% natural gas and oil, 34% nuclear, 5% renewable), solar PV and wind. The study concludes that both steam methane reforming and the auto maintained decomposition alternatives have GHG emissions quite similar to those produced by the wind electrolysis. The GHG emissions from electrolytic processes are about 2.6, 0.59 and 0.3 $kgCO_{2\ eq.}/Nm^3H_2$ for electrolysis based on electricity from grid, solar PV and wind, respectively. However, it should be noted that the CCS storage option may perform worse in other categories due to need for electricity, as discussed also in [Dufour et al., 2009].

[Geerken et al., 2004] performed a review of several LCA studies and well-to-wheel studies on production and use of hydrogen in various stationary and mobile applications. However, they have not provided a critical review on LCA itself; rather this report is a compilation of mentioned literatures. The reason has been given as the final results from different studies with different assumptions are often not easy to compare, because of differences in scope, chosen impacts, reference year of technology, geographical differences, system boundaries, estimated life time of components, etc. This fact has been true also for this current study, even if an attempt has been made for comparison at the end of this review.

[Granovskii et al., 2006] examined various hydrogen production methods (from gasoline, natural gas, and electrolysis via wind and solar) and use of hydrogen in fuel cell vehicles to compare them with a base case (gasoline use in an internal combustion engine). Their analysis included the energy demand and GWP. Like the other mainstream literatures, they concluded that the use of wind power to produce hydrogen via electrolysis, and its application in a fuel cell vehicle, is characterized by the lowest greenhouse gas emissions and fossil fuel energy consumption. However, the economic attractiveness of wind technology depends significantly on the ratio in costs for hydrogen and natural gas. At a cost ratio of 2, capital investments to produce hydrogen via natural gas are about five times lower than to produce hydrogen via wind energy. They further conclude that solar electrolysis is advantageous by resulting in less air pollution compared to natural gas reforming method. Corresponding emissions for natural gas SMR, solar and wind electrolysis are reported to be about 85, 30 and 20 $gCO_{2\ eq.}/MJH_2$ (based on LHV), respectively.

[Hacatoglu et al., 2012] made a study on comparative life cycle assessment of hydrogen production between nuclear based thermochemical water decomposition and water electrolysis via wind and solar electricity. Their results show that the GHG emission via nuclear based thermochemical pathway is about 27 $gCO_{2\ eq.}/MJH_2$, out of which over one third is from fuel (Uranium) production stage that needs a larger amount of coal as energy source. This emissions value is greater than that via wind based water electrolysis pathway (20 $gCO_{2\ eq.}/MJH_2$). Solar electrolysis performed worse than both, i.e. about 30 $gCO_{2\ eq.}/MJH_2$. Not surprisingly the natural gas and gasoline routes performed the worst, i.e. about 85 $gCO_{2\ eq.}/MJH_2$. This and many other studies have a common limitation that only the global warming potential of the process is quantitatively assessed. Quantitative statements for AP, POCP, EP, etc. are also needed for a better understanding of the processes and their comparison with others.

Also the societal concerns of use of nuclear fuel management and nuclear waste disposal are not taken into consideration before making the conclusions that nuclear based option is far better than natural gas based option.

[Ivy, 2004] has compiled the energy requirement data for electrolysers using the different manufacturer's (e.g. Stuart, Teledyne, Proton, Norsk Hydro, Avalence) electrolytic hydrogen generation system. These electrolysers are all alkaline types except that from Proton which was a PEM electrolyser. The system capacities for the chosen elctrolyser models from these five manufacturers were varying i.e. 60, 42, 10, 485 and 4.6 Nm^3/hr, respectively. The corresponding energy consumption values for the electrolyser system were 4.8, 5.6, 6.3, 4.8 and 5.4 kWh/Nm^3, respectively. This study further discusses the economics of electrolysers. The cost of producing hydrogen via current electrolytic processes is largely dependent on the cost of electricity, the efficiency of the systems, and the capital costs of the systems. The cost of electricity and the system efficiencies are interrelated because either an increase in efficiency or a decrease in electricity costs will bring down the overall electricity cost contribution and thereby unit hydrogen production costs. However, the system efficiency that can be increased is limited for technical reasons. Therefore there is a broad interest to reduce the energy requirement of the system. Exactly to this point, the use of excess electricity generated from renewables would be the best solution not only from environmental viewpoint but also from economical.

[Koroneos et al., 2004] performed a LCA study to compare the environmental impact of hydrogen production methods from high pressure alkaline electrolysis using renewable energy electricity (wind, hydropower, solar PV, solar thermal and biomass) to steam reforming of natural gas. Their study is based on the Global Emission Model for Integrated Systems (GEMIS) database. The examined impact categories are GWP, AP, EP, and winter smog (solid particulate matters, SPM). For all these four categories, electrolysis via wind electricity shows the best results followed by hydroelectricity and solar thermal electricity. High equivalent emissions of CO_2 and SO_2 have the major negative impact on hydrogen production by steam reforming of natural gas. Methane (CH_4) emissions, which primarily come from natural gas losses to the atmosphere during production and distribution, have a large effect on the GWP of the system. This paper further uses the Eco-indicator method to compare the overall impacts of the mentioned technologies as a single score value. Surprisingly, their results for single score show the solar PV be the worst option with the score of 0.05 mPt/MJH_2. This is followed by biomass, natural gas, solar thermal, hydropower, and wind electrolysis, which has the score of less than 0.005 mPt/MJH_2. It is important to note that CO_2 equivalent emission for natural gas option is double to that for PV system (0.08 vs. 0.04 kg/MJH_2), whereas the overall single score point for the former one is only about 0.75% of the later one. This shows the clear risk of considering only one impact category in LCA analysis.

In the other study, [Koroneos et al., 2008] compared two biomass-to-hydrogen systems: biomass gasification by reforming of the syngas, and gasification followed by electricity generation and thereby electrolysis. The emissions in terms of CO_2, SO_2 and PO_4 equivalent have been analyzed. Environmental impact to a single score value is calculated using the Eco-

indicator 95 method. While the gasification-electrolysis route has a greater eutrophication effect, biomass gasification and reforming of syngas route has higher environmental impacts in terms of GWP and AP. In addition, weighting results demonstrate that the biomass-gasification-electricity-electrolysis route has better environmental performance than the process involving reforming of syngas (0.123 vs. 0.263 mPt/MJH$_2$). One of the main reasons for this was the assumption that the biomass-gasification-electricity power plant produces all the electricity required for electrolysis and hydrogen liquefaction steps without need of additional power source for these provisions. On the other hand the gasification steam reforming plant requires additional electricity due to compression requirements that involve the steam reforming and PSA processes. This electricity is assumed to come from the grid, which is mainly fuelled with non-renewable energy sources. The biomass-gasification-electricity-electrolysis route requires 4.2 TJ of primary energy input per TJ of hydrogen, whereas the biomass-gasification-steam reforming-PSA route requires only 2.4 TJ per TJ hydrogen produced. But gasification-electricity-electrolysis route has 93% share of renewable energy in the primary energy input compared to only 54% for its counterpart. Thus the gasification-steam reforming-PSA route is the most energy efficient. Such variations in assumptions make it clearly difficult to compare the overall environmental impacts of one route to the other.

[Lee et al., 2010] studied the environmental and economic aspects of the electrolytic hydrogen generation system using LCA and life cycle costing (LCC) methodologies. The studied H$_2$ production pathways are water electrolysis with wind power and with Korean electricity mix of 2007 (coal: 38%, nuclear: 36%, LNG: 20%, oil: 5%, hydro: 1%, other renewables: 0.2%). In LCA results, wind route has been reported to be superior to the other pathway in GWP and abiotic resource depletion categories. The LCC of the target fuel pathways consist of the well-to-tank (WTT) costs and the tank-to-wheel (TTW) costs. Although grid and wind electrolysis routes incur high capital costs owing to the required capital investment in fuel cell vehicles (FCVs), they have lower well-to-wheel (WTW) costs than those of conventional gasoline fuels due to the high FCV efficiency in fuel utilization stage. Thus, this study concluded that the future competitiveness of wind pathway in both environmental and economic aspects.

[Marquevich et al., 2002] conducted a life cycle inventory analysis to assess the environmental load, specifically GWP, associated with H$_2$ production by steam reforming of feedstock (methane and naphtha) and vegetable oils (rapeseed oil, soybean oil and palm oil). The GWP of H$_2$ produced from rapeseed oil, palm oil and soybean oil are found to be 6.42, 4.32 and 3.30 kgCO$_{2\ eq.}$/kgH$_2$, respectively and the GWPs associated with the production of H$_2$ by methane and naphtha steam reforming are 9.71 and 9.46 kgCO$_{2\ eq.}$/kgH$_2$, respectively. Furthermore, they have calculated the GWP for 20 and 500 years period, the impacts being higher for 20 years and lower for 500 years period. Their conclusion states that GWP can be significantly reduced if natural gas and naphtha are replaced by vegetable oils.

Life cycle approaches of the emerging energy technologies has been documented in NEEDS project report [NEEDS, 2008] under the 6th framework program of EU. This report also com-

piled some properties of different types of electrolysers. The energy consumption values in kWh/Nm³H₂ are given in the range of 4.3-4.9 for conventional alkaline, 3.8-4.3 for advanced alkaline, 3.6-4.0 for PEM and 2.5-3.5 for high temperature SOEs. It report contains relatively detailed LCI data on electrolytic hydrogen production plant. The LCA analysis includes four phases: fuel supply (i.e. electricity needed for electrolysis), production (of electrolyser components and accessories), operation (component replacement, maintenance, etc. excluding electricity supply) and disposal. Fig. 15 shows the contribution analysis for electrolytic hydrogen production by using then UCTE grid mix. A huge contribution in all emission categories comes from electricity supply, meaning from the fuel used to generate the grid electricity. Since a high amount of electricity is required for electrolysis, the source of electricity is very important. The use of renewable energy generated electricity would shift this contribution virtually towards zero.

Fig. 15: Contribution analysis for electrolytic hydrogen production
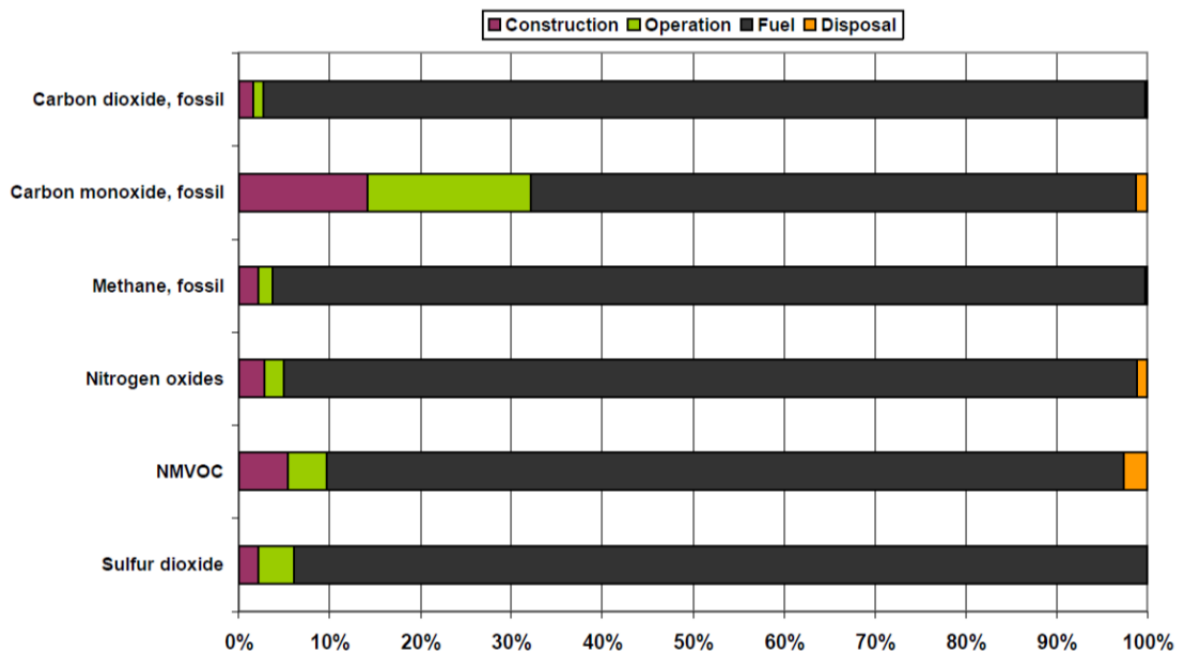
Source: [NEEDS, 2008]   IEK-STE 2012

In [NETL, 2006] the life cycle emissions of greenhouse gases released during the production of hydrogen in the United States from liquefied natural gas (LNG) and coal has been analyzed. For the production of hydrogen from LNG, GHG emissions are considered from all process steps, which include natural gas production, liquefaction, shipping by tanker, re-gasification, pipeline transport, and ultimately conversion to hydrogen via SMR. Life cycle GHG emissions of hydrogen production from LNG plant with CCS provision were compared with the emissions from plant that do not employ CCS. The results show that uncontrolled GHG emissions are 12.4 and 8.9 kgCO₂ eq./kgH₂ for underground mined coal and LNG, respectively. However, when CCS and coal mine methane (CMM) mitigation (during coal mining) measures are employed; the figures would be reduced to as low as 1.9 and 4.7 kgCO₂

$_{eq.}$/kgH$_2$ for surface mined coal and LNG, respectively. CMM mitigation methods are evaluated for reducing methane emissions both by capture and use of concentrated methane and by oxidation of methane in mine ventilation air.

[Ozbilen et al., 2011a] conducted a comprehensive LCA of commercial hydrogen production methods by including all the major process steps involved in every method. By using both CML 2001 and Eco-indicator 95 methods they examined the energy consumption and the carbon dioxide equivalent emissions of each production method. Five different technologies were investigated for a location in Toronto - steam reforming of natural gas, coal gasification, water electrolysis by wind and solar electricity, and thermochemical water splitting with Cu-Cl cycle. The results showed that steam reforming of natural gas has the highest environmental impact, whereas thermochemical cycles have the lowest. It should be noted that the study considered only GWP and AP. The paper's results are summarized in Tab. 5.

Tab. 5: Environmental impacts of H2 production methods

H$_2$ production method	GWP (gCO$_2$ $_{eq.}$/kgH$_2$)	AP (gSO$_2$ $_{eq.}$/kgH$_2$)
Nuclear based Cu-Cl cycle	737	1.75
Nuclear based S-I cycle	411	2.41
Nuclear based high temperature electrolysis	2,000	4.84
Natural gas steam reforming	12,000	14.52
Biomass based electrolysis	3,000	29.03
Wind based electrolysis	1,200	2.58
Solar based electrolysis	2,000	8.07

Source: [Ozbilen et al., 2011a]   IEK-STE 2012

The acidification potential results show that biomass gasification has the highest impact on environment followed by high temperature electrolysis. No further elaboration has been made. Nuclear based electrolysis also has higher AP. Even though operation of a nuclear plant does not emit CO$_2$, there is a significant GWP contribution of using this option. In a nuclear plant, the nuclear fuel cycle (mining, fabrication and transportation) accounts for 63% of the total CO$_2$ emissions; whereas materials, construction, operation and waste disposal contribute for 5%, 8%, 15%, and 9%, respectively. In order to measure the overall environmental impacts, the Eco-indicator 95 method has been applied. It has been then concluded that natural gas steam reforming has the highest overall environmental impact. Hydrogen production processes via nuclear based thermochemical cycles and renewable ener-

gy based electrolysis have much lower effects on the environment compared to via natural gas steam reforming. Nuclear based S-I cycle is presented to be the winner among the studied technologies and Cu-Cl cycle follows this cycle. Wind electrolysis has almost double impacts than that from S-I cycle. Nuclear based high temperature electrolysis and solar based electrolysis have similar impacts, both being about 50% higher than wind based electrolysis. Impact from biomass based electrolysis is about double than that from wind electrolysis. These authors have published several other papers about the LCA of hydrogen production, focusing on nuclear based thermochemical water splitting cycles [Ozbilen et al., 2011b], [Ozbilen et al., 2012a], etc. In one of their paper, i.e. [Ozbilen et al., 2012b], they have also analyzed the abiotic resource depletion potential and radiation impacts. However, these results are only presented for nuclear Cu-Cl cycle and no comparison has been made with the other methods, leading no overall conclusions on choosing the best technology.

[Solli et al., 2006] presented a comparative hybrid LCA (combining process LCA and input-output model) to evaluate two different methods for hydrogen production. The environmental impacts of hydrogen production from nuclear based water splitting are compared with those from natural gas steam reforming with CCS. For the impact assessment, the CML 2000 baseline method and the Eco-indicator 99 method are used. Functional unit has been set as 1 TJH_2 (HHV). The natural gas based system performs better for HTP and radiation (RAD) whereas the nuclear alternative has a better score for GWP, AP and EP; the impacts often associated with emissions from combustion. Except for radiation, the numbers are in the same order of magnitude, ratio from natural gas to nuclear varying from factor 1.4 to 4.5. They have not applied a weighting procedure, hence no overall winner has been proclaimed. This is one of the few studies assessing the HTP and RAD categories. Uranium mining and milling are the most important contributions to HTP impact category, alone accounting for more than 60% of impacts. Open pit mining is significantly better than underground mining in this impact category. The radiation impacts from the nuclear system are dominated by the mill tailings that continue to exhale radon for thousands of years. Improved management of mill tailings is clearly the most important strategy for reducing the human health impacts from ionizing radiation.

Also referenced by almost all authors publishing papers on wind based electrolysis, GWP data for hydrogen produced by natural gas steam reforming and by wind/electrolysis are reported by the National Renewable Energy Laboratory [Spath & Mann, 2001] and [Spath & Mann, 2004]. [Spath & Mann, 2001] performed LCA of hydrogen production via natural gas steam reforming to examine the net emissions of greenhouse gases and the other major environmental consequences. Material and energy balances are performed in a cradle-to-grave manner on the operations required to transform raw materials into useful products. Natural gas lost to the atmosphere during production and distribution is also taken into account. The overall GWP of the system is about 12 $kgCO_2$ $_{eq.}$/kgH_2. CO_2 is the main contributor, accounting for 89.3% of the GWP for this specific system, followed by methane (10.6%) and N_2O (0.1%). Hydrogen plant operation contributed about 75% of this value, while the rest being from natural gas production and transport; construction and decommissioning of

plant; electricity supply to the system for purification; etc. In terms of resource consumption, as anticipated, natural gas is consumed at the highest share, accounting for 94.5% of the total resources on a weight basis, followed by coal 4.1%, iron (ore plus scrap) 0.6%, limestone 0.4%, and oil 0.4%. Water consumption at H_2 plant was about 19 ltr/kgH$_2$. The size of the hydrogen plant was about 1.5 million Nm^3/day.

[Spath & Mann, 2004] performed a life cycle assessment on a renewable hydrogen production process which employs wind-electrolysis. Resource consumption, and energy use, material and energy balances are performed in a cradle-to-grave manner. For the wind/electrolysis system, the material production processes required to construct the wind turbines, electrolyser, and hydrogen storage tanks were taken into account. The system incorporates three 50 kW wind turbines with a 30 Nm^3/hr electrolyser. This electrolyser converts the electricity to hydrogen with an efficiency of 85% (HHV). The product hydrogen is compressed to a pressure of 20 MPa, stored, and dispensed at the fueling station.

Fossil fuels, metals, and minerals are used to produce hydrogen in this process. The iron, which is mostly used in manufacturing the wind turbines and hydrogen storage vessels, accounts for 37.4% of the resources used. The large amount of limestone, 35.5% of the major resources, is used for the turbines' concrete foundations. Coal, which is consumed primarily to produce the steel, iron, and concrete, accounts for 20.8% of the remaining resources. This is followed by oil at 4.7%, and natural gas at 1.6% which are primarily used in manufacturing the wind turbines. Water is consumed not only in the electrolysis operation, but also in upstream processes. For each kg of hydrogen produced, 26.7 liters of water are consumed by the system. Nearly 45% is used by the electrolyser, while 38% and 17% is used in manufacturing wind turbines and hydrogen storage vessels, respectively. The average resources consumed in the process are summarized in Tab. 6 (percentage numbers are rounded).

Tab. 6: Average resource consumption in wind electricity electrolysis system

Resource	Total (g/kgH$_2$)	From wind turbines (%)	From electrolysis (%)	From storage (%)
Coal	214.7	67	5	27
Iron (Fe, ore)	212.2	64	6	30
Iron scrap	174.2	53	8	39
Limestone	366.6	96	1 (0.3)	3
Natural gas	16.2	72	15	13
Oil	48.3	76	13	11

The GWP is presented to be about 970 $gCO_{2\ eq.}$/kgH_2. About 78% of it was from wind turbine production and operation; about 4.4% was from electrolysis production and operation; and about 17.6% was from hydrogen compression and storage. Average energy consumption was 9.1 MJ/kgH_2. The majority of the energy consumption, i.e. 72.6%, was from manufacturing of the wind turbines (LHV basis). Contribution form electrolysis was 4.8% and that from storage was 31.6%. Tab. 7 shows air emission values (percentage numbers are rounded). CO_2 is emitted at the highest rate, about 95% by weight. In general, the majority of the air emissions come from the process steps in manufacturing the wind turbines.

Tab. 7: Average air emissions from wind based electrolysis

Air emission	Total (g/kgH_2)	From wind turbines (%)	From electrolysis (%)	From storage (%)
Carbon dioxide	950	78	5	17
Carbon monoxide	0.9	80	4	16
Methane	0.3	92	3	5
Nitrogen oxides	4.7	46	47	7
Nitrous oxides	0.05	67	6	27
Non-methane hydrocarbons	4.4	63	7	30
Particulates	28.7	94	1	5
Sulfur dioxide	6.1	62	26	12

[Utgikar & Thiesen, 2006] have performed a life cycle assessment of high temperature electrolysis for H_2 production via nuclear energy (high temperature electrolysis of water vapor). The electrolytic hydrogen production emissions results have been compared with the renewable energy based electrolysis using the results reported by [Koroneos et al., 2004] and [Spath & Mann, 2004]. High temperature electrolysis is advantageous to low temperature alkaline electrolysis because of its higher efficiency, which is due to reduced cell potential and consequent electrical energy requirements. The largest source of CO_2 emissions by life cycle stage in nuclear electricity generation is fuel mining, fabrication, and transportation,

which accounts for 63% of the total CO_2 emissions. Of the total; materials, construction, operation, and waste disposal contribute 5%, 8%, 15%, and 9%, respectively [White & Kulcinski, 2000]. The emissions associated with the production of solid oxide fuel cells are mainly the primary production of iron, nickel, and chromium, accounting for 40% of the global warming and 50% of the acidification impacts. The combined nuclear-HTE plant GWP emissions are reported as 2 kg$CO_{2\ eq.}$/kgH_2. The AP for this system is 0.15 gH^{ion}/kgH_2. The HTE process has lower greenhouse gas emissions and the acidification potential than all other processes described, with the exception of wind and hydropower produced electricity coupled with conventional electrolyser. The steam-reforming process emits nearly six times as much carbon dioxide equivalents and has three times the AP as the HTE process. The environmental impacts (both global warming and acidification) of the nuclear-HTE system are primarily due to the activities associated with mining, fabrication and construction of the plants. These activities will occur over a short time period compared to the actual operation of the plant, which itself has relatively minor adverse impact on the environment. It may be easier to manage these short-term emissions through carbon dioxide sequestration and acid gas neutralization technologies to mitigate their environmental impact.

In their recently published paper, [Wulf & Kaltschmitt, 2012] have analyzed the overall life cycle hydrogen production at a hydrogen refueling station in Germany. In this station at least 50% of hydrogen is produced on-site by water electrolysis from renewable sources (wind). The remaining other 50% of hydrogen is provided by trucks from a large-scale production plant where hydrogen is produced from glycerol as a by-product of the biodiesel production. These two pathways are compared with hydrogen production from natural gas steam methane reforming, and biomass and coal gasification. Their results show that the hydrogen production based on water electrolysis fed by German grid electricity mix of 2010 (16% renewable, 22% nuclear and rest fossil) should be avoided to reduce GHG emissions from life cycle perspective. Steam methane reforming of natural gas is better than grid based electrolysis in terms of GHG reduction because of efficiency losses in fossil fuel – electricity – electrolysis route. The major emissions from the hydrogen production by electrolysis are caused by electricity generation, i.e. up to 99.8% from German grid and up to 96.8% from green electricity. The remaining tiny amount is caused by the production of the electrolyser itself and the potassium hydroxide needed to operate the system. As expected the lowest emissions can be achieved by the electrolysis with green electricity followed by the gasification of wood biomass. The GWP values are 32, 4.1, 4.1 and 12.9 kg$CO_{2\ eq.}$/kgH_2 for grid electricity electrolysis, green electrolysis, wood gasification and steam methane reforming, respectively. However, other environmental impacts may have to be analyzed, e.g. land use change if biomass wood gasification is used. The emissions from pyro-reforming of glycerol are unexpected high, similar to that of coal gasification (about 23 kg$CO_{2\ eq.}$/kgH_2) even though a renewable resource is used. The reason for that is the emission caused during the production of glycerol (in particular the overall pre-chain up to the agricultural production of the rape seed).

V   Conclusions

Several hydrogen production methods are discussed in the literature. They range from fossil fuel based systems such as natural gas steam methane reforming to renewable energy based systems such as wind based electrolysis. Other advanced types, including those based on nuclear energy, are also presented as future potential technologies. However, until today almost all the industrial hydrogen is produced by using fossil fuels. Steam methane reforming has been the most widely used method. Production of hydrogen has been so far mainly for other purposes than energetic use. Major consumers are chemical industries such as ammonia production and oil refineries. It is expected that the energetic use of hydrogen will be speeding up in the future, beginning with the mobile applications - fuel cell and direct combustion vehicles. Use of renewable energy for water electrolysis could be one of the best methods for producing hydrogen from ecological perspectives.

Use of renewable energy based hydrogen production technologies is not always environmentally neutral. During the operational phase, where energy is supplied via renewable sources, the process may be carbon neutral; however, the manufacturing and installation of renewable energy plant is associated with certain emissions. A LCA is a scientifically valid and accepted tool to perform the environmental analysis of certain process or technology. The results could be used to identify the weak points in process chain as well as to compare one process to another.

In this study, different literatures discussing the environmental LCA of hydrogen production technologies have been reviewed. System boundary of these studies varies mainly after the hydrogen production phases, e.g. storage or dispensing, supply as fuel to vehicles, supply to fuel cell, etc. The functional unit also varies much. The mostly used unit is kgH_2. Other units in use are Nm^3, MJH_2, etc. The impact assessment method used is largely the CML method. However, the eco indicator method is also used by some authors. Some use both methods to analyze the same data. The following conclusions have been drawn from this review:

- Several studies are published on hydrogen production techniques. Major hydrogen production methods are steam methane reforming of natural gas followed by coal gasification. Share of electrolysis is still small, i.e. about 4%.

- Three types of electrolysis are discussed in the literature: alkaline, polymer membrane electrolyte, and solid oxide electrolysis. However, LCA studies discuss mainly alkaline electrolysis.

- Most of the LCA studies compare environmental impacts of electrolytic hydrogen production methods with conventional methods.

- Some LCA studies dealing with electrolysis compare the environmental impacts of hydrogen production via conventional grid electricity with via renewable energy (mainly wind and solar) electricity.

- Global warming potential is analyzed by almost all the authors and the values vary from one to the other study as shown in dark blue color in Fig. 16 (e.g. value for

steam methane reforming of natural gas varies from 8.9 to 12.9 kgCO$_2$ $_{eq.}$/kgH$_2$). The second category discussed is acidification potential (Fig. 17). The rest categories are not often mentioned. However, some consider the primary energy demand; others mention resource use including the water consumption in electrolytic process. One study mentioned solid particulate matters and the other did human toxicity and radiation impacts of using nuclear based water splitting methods.
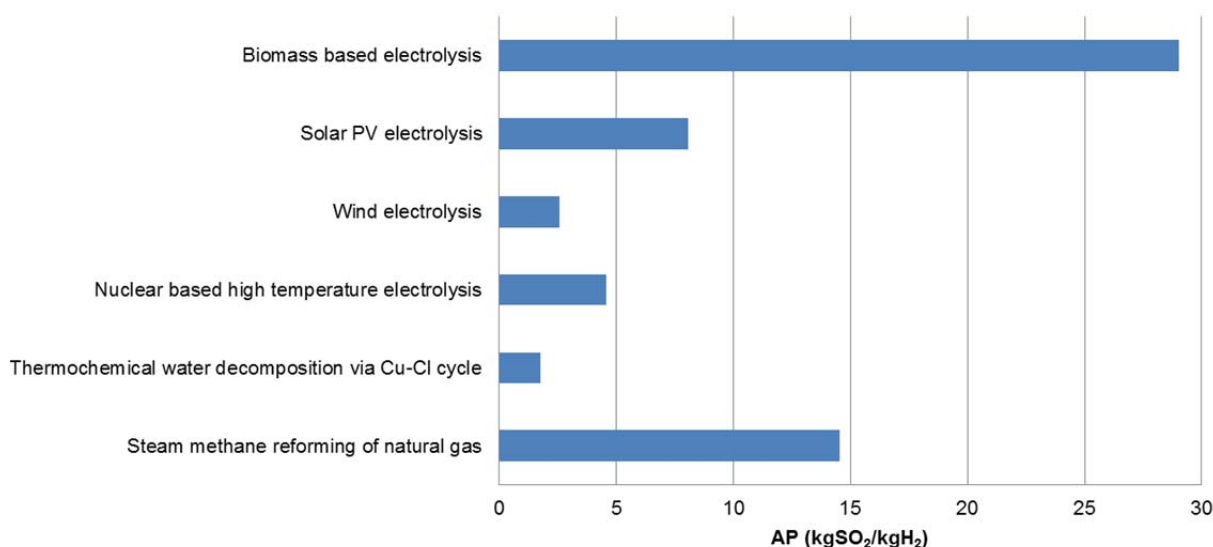
Fig. 16: A range of GWP for different hydrogen production methods
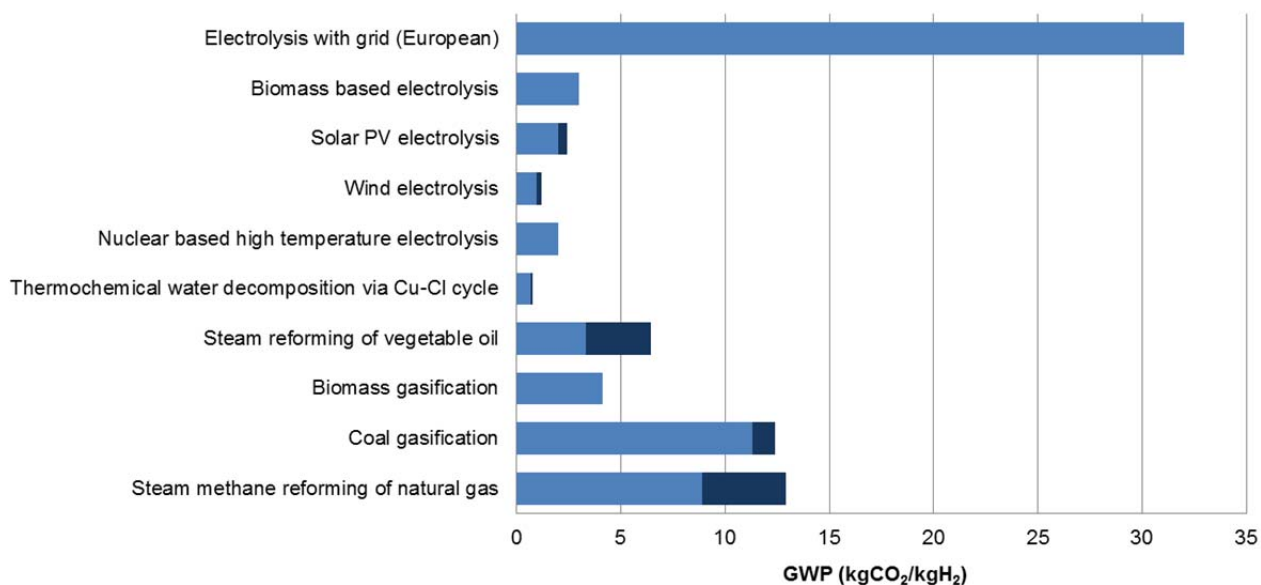
Source: [Bhandari, 2012] (compilation from reviewed papers)   IEK-STE 2012

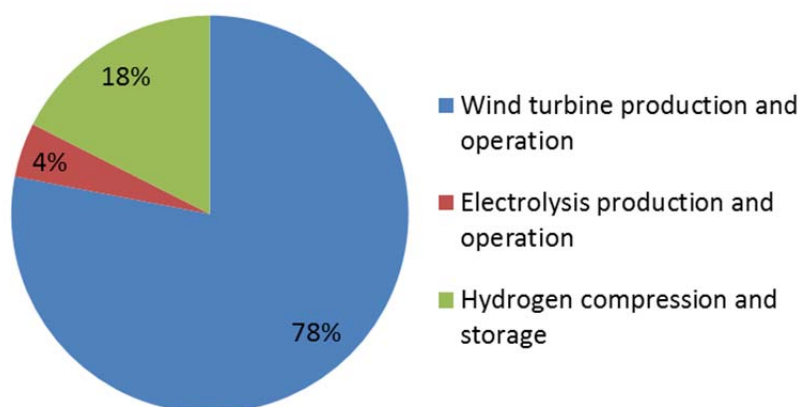**Fig. 17: A range of AP for different hydrogen production methods**

Source: [Bhandari, 2012] (compilation from reviewed papers)   IEK-STE 2012

- As shown previously also in Fig. 15, the environmental concern of electrolytic process for hydrogen production is mainly in its operation phase – i.e. electricity supply (based on fossil fuel or renewable resources generated).

- Not all the studies work with primary data. In many cases the primary data are cited repeatedly, e.g. for wind based electrolysis [Spath & Mann, 2004] is the major reference cited by many authors. For nuclear water splitting, design data has been used, since there are not yet commercially operational units in the market.

- LCA of electrolytic H_2 production using wind energy shows that adverse environmental contribution from electrolyser is relatively small, i.e. only 4% (Fig. 18). In contrast, the wind turbine itself is the major contributor to GWP and other impact categories.

Fig. 18: Share of GWP (970gCO$_2$ eq./kgH$_2$) in wind electrolysis

- 18%
- 4%
- 78%
- Wind turbine production and operation
- Electrolysis production and operation
- Hydrogen compression and storage

Source: data from [Spath & Mann, 2004]   IEK-STE 2012

- Electrolyser has been analyzed as a single component. Therefore details on the contribution from individual components of electrolyser such as electrodes or membrane (asbestos) could not be understood from this review. A separate and electrolyser focused LCA study needs to be carried out to understand individual component's contribution.

In future, it is recommended to broaden the scope of the LCA studies on hydrogen production methods by including other impact categories beyond the GWP and there is a need for a more exposure orientated assessment of the potential impacts on human health and ecology. It would only be possible to compare the different electrolytic methods (alkaline, PEM and SOE) from LCA perspective if the electrolyser is not analysed as a single component of hydrogen production process chain, rather the contribution from individual components of the electrolyser are also detailed. Unless such data are available, it will not be possible to compare the environmental impacts of membranes used in the electrolysers, i.e. asbestos vs. polymer membranes.

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